Evaluation of Partial Oxidation Fuel Cell Reformer Emissions

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Objectives

- Measure the emissions from a partial oxidation/autothermal reformer (POx/ATR) fuel processor for a proton exchange membrane fuel cell (PEMFC) system under both cold-start and normal operating conditions
- Assess the feasibility of meeting emissions standards for automobiles and light-duty trucks through the use of a fuel cell vehicle with a flexible-fuel reformer

Technical Barriers

This project addresses the following technical barriers from the Fuel Cells section of the Hydrogen, Fuel Cells and Infrastructure Technologies Program Multi-Year R,D&D Plan:

- K. Emissions and Environmental Issues

Approach

- Define a representative test cycle consisting of both startup and normal operating conditions
- Use the established test cycle to quantify emissions from a POx reformer before and after anode gas burner (AGB) treatment
- Measure emissions with continuous emissions monitoring (CEM) measurements supplemented with laboratory analyses of speciated hydrocarbons and particulate matter (PM)
- Use reasonable approximations and estimates to convert emissions data from a grams/unit fuel basis to a predicted grams/mile basis

Accomplishments

- Measured emissions from a gasoline-fed fuel processor (without fuel cell) over several operating conditions
- Measured emissions from an ethanol-fed fuel processor with fuel cell over several operating conditions (analysis pending)
- Speciated total hydrocarbon (THC) data before and after the AGB
- Assessed the sensitivity of monitoring equipment over a range of operating conditions
- Analyzed data to report emissions on a g/kg fuel basis

Future Directions

- Perform extensive emissions testing of a fuel cell/reformer system to include particulate, formaldehyde, and ammonia as well as NO_x, hydrocarbons, and CO
- Sample and analyze emissions from Nuvera Fuel Cells fuel processor-fuel cell system in 2003
- Project on-road emissions from fuel cell vehicles with on-board reformers

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Introduction

Fuel reformer operation is generally divided into two operating modes: startup and normal partial oxidation. During startup, the fuel processor burns fuel at near stoichiometric conditions until critical system temperatures and pressures stabilize to target values. Once the target conditions are reached, the reformer operates in normal mode in which the fuel processor burns fuel at very rich conditions. Since these modes are comprised of considerably different operating conditions, it follows that the emissions associated with each of these modes are also considerably different.

The combustor is typically cold under startup conditions, generating emissions during this brief period (target times are under 30 seconds) that can be substantially higher than those produced during the remaining, much longer portion of the driving cycle. The pollutant emissions produced during startup operation include NO_x, CO, formaldehyde, and organic compounds. These organic compounds, including hydrocarbons, alcohols, and aldehydes, are regulated in California and referred to as non-methane organic gases (NMOG). Under normal, fuel rich operating conditions, virtually no NO_x is formed, although the formation of ammonia is possible. Most hydrocarbons are converted to carbon dioxide (or methane and/or hydrogen if the reaction is incomplete); however, trace levels of hydrocarbons can pass through the fuel processor and fuel cell. The shift reactors and the preferential oxidation (PrOx) reactor reduce CO in the product gas, with further reduction in the fuel cell. Thus, of the criteria pollutants (NO_x, CO, and hydrocarbons [NMOG]), NO_x and CO levels are generally well below the most aggressive standards. NMOG concentrations, however, can exceed emissions goals if these are not efficiently eliminated in the catalytic burner.

Approach

In this study, a gasoline fuel processor and an ethanol fuel processor were operated under conditions simulating both startup and normal operation. Emissions were measured before and after the AGB in order to quantify the effectiveness of the burner catalyst in controlling emissions. The emissions sampling system includes CEM for O_2, CO_2, CO, NO_x, and THC. Also, integrated gas samples are collected in evacuated canisters for hydrocarbon speciation analysis via gas chromatography (GC). This analysis yields the concentrations of the hydrocarbon species required for the California NMOG calculation. The PM concentration in the anode burner exhaust is measured through the placement of a filter in the exhaust stream.

Emissions data will be used to project on-road emissions for fuel cell vehicles with reformers. Emissions data will be characterized in terms of startup or reforming modes. Although current fuel processor technologies are not configured to follow a typical vehicle load profile, hybrid vehicle power management strategies may facilitate using such fuel processors. For this project, the fuel processor is operated at several steady-state points while emissions are monitored for the steady-state conditions and transients between load changes. The data collected during startup, different loads, and transients serve as inputs to a vehicle emissions model. Using these data, this vehicle emissions model then predicts the emissions for each second in a driving cycle based on load. Startup emissions are considered along with the total driving emissions.

Results

During FY 2003, we performed two series of emissions tests: one gasoline ATR system tested without a fuel cell and one ethanol ATR system tested with a fuel cell. The emissions tests involved sampling criteria pollutants and CO_2 from each of these systems at various operational load points, as described in the previous section. Concentrations of the aforementioned species are obtained using the emissions sampling system shown in Figure 1. Although emissions data have been collected for both the gasoline and ethanol systems, only the gasoline system data have been sufficiently analyzed to allow presentation of results at this time.

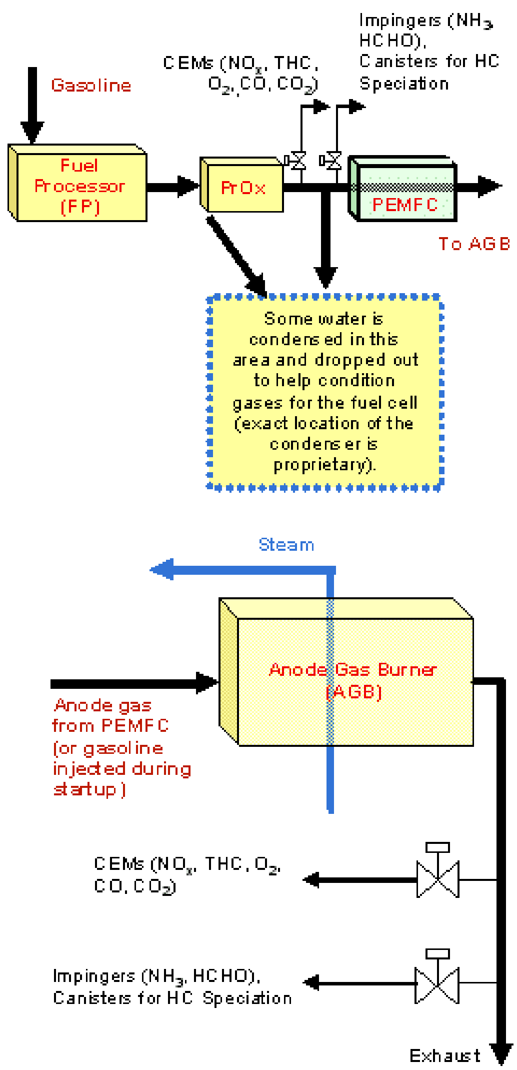

Figure 1. Emissions Testing Setup for the Fuel Processor and Fuel Cell System

In general, testing did not attempt to follow a vehicle driving cycle, but rather followed a series of steady-state conditions with load changes. Figure 2 shows emissions levels at the AGB for the beginning of a gasoline reformer testing series. This series of operating load points was started at low load conditions after a hot start (about 6 kWe or 12% load) and brought to steady state at 12 kWe. THC emissions at the AGB outlet were below the monitor detection limit throughout the testing period. CO emissions were highest at the lower load points and dropped when the power was increased above 10 kWe, or 20% full power. NO_x emissions stabilized at about 20 ppm for operation above 10 kWe.

In order to determine how current emissions performance would compare to existing vehicle standards, these emissions levels were converted to a gram per mile basis using the system power input and typical vehicle efficiencies. The results of this conversion are given in Figure 3 along with the corresponding California super ultra low emission vehicle (SULEV) passenger vehicle standards. For the series shown in Figure 2, CO and THC emissions are much lower than the SULEV standard. For the same test series, the NO_x emissions are on the same order of magnitude as the SULEV standard, indicating that the reformer system may require additional optimization to meet the SULEV NO_x standard.

The PM concentrations for both tests correspond to emissions levels below California SULEV vehicle emissions standards. These PM concentrations were determined by passing the entire exhaust stream

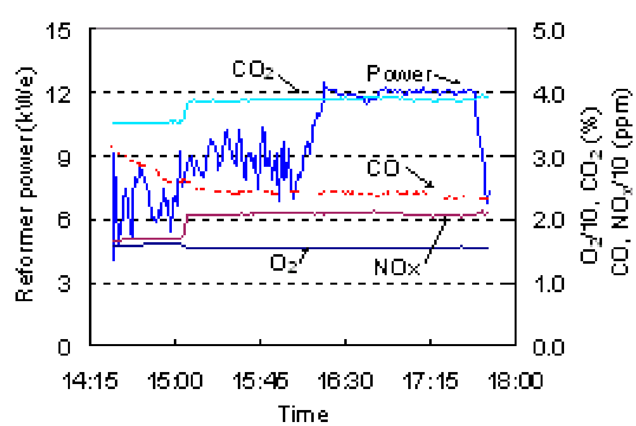

Figure 2. CEM Emissions Test Results at the AGB Outlet (partial)

Species	AGB* (g/mile)	CA SULEV (g/mile)
NOx	0.05	0.02
CO	0.004	1.0
NMOG**	<0.001**	0.01

*The AGB emissions were not optimized for vehicle emissions. This comparison is to show how current emissions performance would compare to existing vehicle standards.

** THC emissions are given for the AGB. The CA SULEV standard shown is for NMOG, a subset of THC. Both AGB NMOG and THC emissions are lower than the corresponding SULEV standard.

Figure 3. Projected Vehicle Emissions at the AGB Outlet (unoptimized)

	PM (μg/m³)	PM (g/mile)
CA SULEV Standard		0.01
Gasoline ATR system: AGB Test 1	30	<< 0.01
Gasoline ATR system: AGB Test 2	68	<< 0.01

Figure 4. PM Emissions Results at the AGB Outlet

through the PM filter and averaging the integrated PM sample over the total flow that passed through the filter. Figure 4 shows PM concentrations at the AGB determined for steady-state gasoline ATR system operation and the corresponding SULEV standard.

Conclusions

The combustion of reformer products at the AGB did not represent an optimized fuel cell vehicle configuration. Even with these limitations, the following conclusions can be drawn from the data.

- NO_x emissions from an AGB averaged about 20 ppm for the load points tested. These emissions levels would correspond to approximately 0.05 g/mile of NO_x for operation. In order to meet stringent emissions standards, further optimization will be required. Also, other power loads will need to be tested to ensure that emissions are acceptable for all operating modes.

- In-use THC and CO emissions were under 0.2 ppm and 3 ppm, respectively, which would correspond to on-road emissions well below the SULEV standards. More data on startup with an optimized fuel processor is required before startup on-road NMOG and CO emissions can be estimated.